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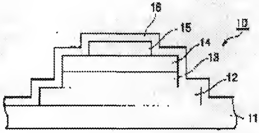
(54) MANUFACTURING METHOD OF ELECTROLUMINESCENT ELEMENT

(57)Abstract:

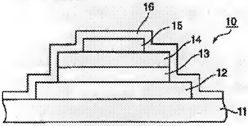
PROBLEM TO BE SOLVED: To improve adhesion and gas barrier characteristics of a protection film.

SOLUTION: With the organic EL element 10 made by laminating a positive electrode 12, an organic hole injection layer 13, an organic light emitting layer 14, a negative electrode 15 and a DLC film 16 on a transparent substrate 11, an RF power impressed on the substrate is increased continuously when the DLC film 16 is formed as a protective film by a plasma CVD method, and inner stress of the DLC film 16 is controlled to be larger as it is directed from the inside of the film toward its surface. That is to say, the film is formed so that the more toward the side each layer of the element is formed, the smaller inner stress the film has, and the more toward the surface side of the DLC film, the larger inner stress the film has.

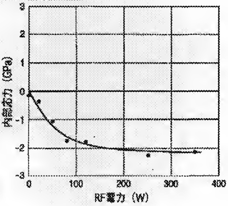


DRAWINGS

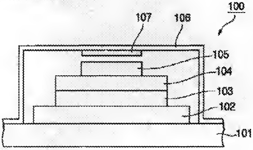
[Drawing 1]



[Drawing 2]



[Drawing 3]



[Translation done.]

DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]
[Drawing 1]It is an outline sectional view showing the laminated structure of the organic EL device shown as an embodiment of the invention.
[Drawing 2]When forming the DLC film of the organic EL device, it is a mimetic diagram showing change of the internal stress of the DLC film at the time of changing the RF power to impress.
[Drawing 3]It is an outline sectional view showing the laminated structure of the organic EL device of structure conventionally.
[Description of Notations]
10 Organic EL device
11 Transparent substrate
12 Anode electrode
13 An organic hole injection layer
14 Organic luminous layer
15 Cathode electrode
16 DLC film

[Translation done.]

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention]This invention relates to the manufacturing method of the electroluminescent element which reconciled the adhesion force and the gas barrier characteristic in a DLC film as a protective film of an electroluminescent element in more detail about the manufacturing method of an electroluminescent element.
[0002]
[Description of the Prior Art]The electroluminescent element (EL element) is also called electroluminescence devices.
Now, the inorganic EL element using the inorganic material as fluorescence material and the organic EL device using organic materials are used.
Among these, the organic EL device is considered as the composition which put the thin film which makes a fluorescence organic compound a subject by the anode and the negative pole.
It is a light emitting device which emits light using discharge (fluorescence and phosphorescence) of the light at the time of making an exciton (EKISHINTON) generate and this being deactivated by making an electron and an electron hole pour in and recombine with this thin film.

[0003]Fundamentally, the organic EL device is constituted by inserting the laminated-structure-ized organic fluorescent substance thin film which consists of organic compounds between the metal electrode which functions as the negative pole, and the transparent electrode which functions as the anode. Here, the thing of the two-layer structure which arranged the organic hole injection layer in contact with the organic luminous layer as a laminated-structure-ized organic fluorescent substance thin film, the thing of the three-tiered structure which put the organic luminous layer with the organic electronic injection layer and the organic hole injection layer, etc. are known.
[0004]By the way, in an EL element, when an organic fluorescent substance thin film is formed on a transparent substrate and forms a transparent electrode in this transparent substrate side, it is considered as the structure which takes out light from this transparent substrate side in many cases. At this time, it is necessary for the upper layer of an organic fluorescent substance thin film to form the

protective film which protects this organic fluorescent substance thin film.

[0005]Conventionally, as shown in drawing 3, EL element 100 of structure on the transparent substrate 101 which comprises glass or a resin material The transparent electrode layer 102, It is closed by the cover material 106 which the organic hole injection layer 103, the organic luminous layer 104, and the cathode electrode layer 105 are laminated one by one, and becomes, for example, comprises a glass material and a metallic material. Inside the cover material 106, it is the purpose of preventing other each class from deteriorating by a steam, oxygen, etc. in the atmosphere, and the moisture absorption material 107 formed, for example of silica gel, BaO, etc. is allocated.

[0006]

[Problem(s) to be Solved by the Invention]By the way, conventionally, since the transparent substrate 101 and the cover material 106 were formed with the glass material or the metallic material, EL element 100 of structure had the problem that flexibility could not be given and it could not form enabling free bending. Although closing by the protective film which comprises DLC (Diamond Like Carbon) as substitution of the cover material 106 of EL element 100 formed on the flexible substrate formed with plastic material etc. is also proposed, The technique of fully securing the both sides of the adhesion force of a DLC film to an element and a flexible substrate and the high gas barrier characteristic is not established.

[0007]Then, the purpose of this invention is as follows.

Be made in view of the conventional actual condition mentioned above, and raise the adhesion force of the DLC film as a protective film.

Provide the manufacturing method of the electroluminescent element which can secure the high gas barrier characteristic.

[0008]

[Means for Solving the Problem]A manufacturing method of an electroluminescent element concerning claim 1 of this invention, It faces forming a DLC (Diamond Like Carbon) film as a protective film with plasma CVD method on an electroluminescent element by which thin film forming was carried out on a substrate, It controls to become large as RF power impressed to the above-mentioned substrate is increased continuously and internal stress distribution of the DLC film concerned goes in a membrane surface from an inside of a film during membrane formation of the above-mentioned DLC film.

[0009]According to the manufacturing method of an electroluminescent element concerning claim 1 of this invention constituted as mentioned above, internal stress distribution of a DLC film can be changed continuously, and stress concentration to specific lamination interfaces can be prevented. For this reason, while being able to raise adhesion force of a DLC film and being able to prevent exfoliation of a film, it becomes possible to secure the high gas barrier characteristic.

[0010]As for variation width of RF power impressed to the above-mentioned substrate, it is desirable for the above-mentioned DLC film to carry out with a maximum RF power value within the limits of conditions which serve as a sp3 joint subject. Thereby, high adhesion force and the good gas barrier characteristic are certainly securable. It is still more desirable to set to 120W the maximum of RF power impressed to the above-mentioned substrate. Thereby, high adhesion force and the good gas barrier

characteristic can be secured still more certainly. In order to raise the gas barrier characteristic further, in a culmination which forms the above-mentioned DLC film, it is desirable to set constant RF power impressed to the above-mentioned substrate. The above-mentioned DLC film can obtain adhesion force in which it is desirable still higher to form on a ground film which comprises any one material of a semiconductor material or an insulating material, and the high resistance metallic material.

[0011]

[Embodiment of the Invention]Hereafter, an embodiment of the invention is described in detail, referring to drawings. Below, when producing the organic EL device 10 as shown in drawing 1, the case where this invention is applied is explained. Then, the laminated structure of the organic EL device 10 is explained first.

[0012]The organic EL device 10 is provided with the following.

The flexible transparent substrate 11 formed with the resin material as shown in drawing 1.

The anode electrode 12 formed on this transparent substrate 11.

The organic hole injection layer 13 formed in the predetermined field on this anode electrode 12.

The organic luminous layer 14 formed on the organic hole injection layer 13, and the cathode electrode 15 formed on the organic luminous layer 14.

As the whole element is covered, DLC film 16 as a protective film is formed in the top layer side of this laminated structure.

[0013]An electron hole transporting bed may be made to intervene on the organic hole injection layer 13 in addition to the laminated structure mentioned above. For example, an electronic injection layer may be made to intervene between the organic luminous layer 14 and the cathode electrode 15.

[0014]In this organic EL device 10, the anode electrode 12 is formed using oxide transparent electrode materials, such as indium oxide, tin oxide indium, and zinc oxide system material, for example.

[0015]the organic hole injection layer 13 is a layer allotted in order to raise the pouring nature of an electron hole to the organic luminous layer 14, and its ionization energy is small enough -- the electron to the organic luminous layer 14 -- shutting up (energy barrier) -- it is formed with a possible material. As such a material, the material of an amine system can be mentioned, for example.

[0016]As a material which forms the organic luminous layer 14, for example JISUCHIRIRU arylene (DSA) system material, Oxadiazole system material, pyrazolo quinoline material, benzooxazol system material, BENZA thiazole system material, benzimidazole system material, a metal chelate-ized oxy NOIDO compound, etc. can be mentioned.

[0017]As a material which forms the cathode electrode 15, alkali system metallic materials, such as Mg-Ag and aluminum-Li, the boron compound which has conductivity, etc. can be mentioned. As a boron compound, the Howe-ized lantern (LaB_6) etc. can be mentioned, for example.

[0018]When an electric field predetermined in the organic EL device 10 constituted as mentioned above to between the anode electrode 12 and the cathode electrodes 15 is impressed, an electron hole is poured into the valence electron level of the organic luminous layer 14 from the anode electrode 12, and an electron is poured in from the cathode electrode 15 to the conduction level of the organic luminous layer 14. Then, an electron hole and an electron move and recombine both the levels in the organic

luminous layer 14. And if these electron holes and an electron are eased by the ground state from an excitation state, the organic electroluminescence nature compound which constitutes the organic luminous layer 14 will emit light.
[0019]Since the transparent substrate 11 is formed with the resin material which has a light transmittance state at this time, this transparent substrate 11 can take out luminescent light from the near field (visual recognition side) which attends the method of outside.
[0020]When producing the organic EL device 10 explained above, each class is formed one by one on the transparent substrate 11, and, finally DLC film 16 is formed. When forming this DLC film 16, the RF power impressed to the transparent substrate 11 is continuously increased using plasma CVD method, and it controls to become large as the internal stress distribution of this DLC film 16 goes to a membrane surface from the inside of a film.
[0021]Here, it faces forming DLC film 16 with plasma CVD method, and the relation between the RF power at the time of changing RF power and the internal stress of DLC film 16 is shown in drawing 2. It turns out that the internal stress of DLC film 16 becomes large, so that the RF power to impress is increased, as shown in drawing 2. Therefore, by increasing the RF power impressed during membrane formation of DLC film 16, the internal stress by the side of the element of DLC film 16 becomes small, and internal stress becomes large as it goes to the surface of DLC film 16. That is, by increasing RF power continuously at the time of membrane formation, as the internal stress of DLC film 16 goes to a membrane surface from the inside of a film, and it becomes large, membranes can be formed.
[0022]Thus, since it will be in the state where internal stress is small, by the interface of a 15 or less-cathode electrode element and DLC film 16 by stopping RF power low in the membrane formation initial stage of DLC film 16, the stress added to the organic EL device 10 whole can be reduced. Therefore, the reliability of an element improves. Since the internal stress of DLC film 16 is changing continuously, the stress concentration to specific lamination interfaces can be prevented, and exfoliation of DLC film 16 can be prevented.
[0023]As for the variation width of RF power, it is desirable for DLC film 16 to carry out with a maximum RF power value within the limits of the conditions which serve as a sp3 joint subject. More specifically, it is [passage clear from drawing 2] desirable to set the maximum of the RF power to impress to 120W. The adhesion force of DLC film 16 can be heightened by this, simultaneously the high gas barrier characteristic can be secured.
[0024]In order to raise further the gas barrier characteristic in DLC film 16, in the culmination which forms DLC film 16, it is desirable to set constant RF power to impress.
[0025]DLC film 16 can obtain the adhesion force in which it is desirable still higher to form on the ground film which comprises any one material of a semiconductor material or an insulating material, and the high resistance metallic material.
[0026]
[Example]Below, based on the organic EL device 10 made into the structure mentioned above, the result obtained when DLC film 16 was actually formed is explained.
[0027]First, on the glass substrate in which the ITO electrode (transparent electrode 12) which patterned after predetermined shape was formed, the organic electroluminescence film (the electron hole

transporting bed 13 and the organic luminous layer 14) was formed with the vacuum deposition method, and LiF/Al film was further formed as the cathode electrode 15.

[0028]Next, the ground film which uses a sputter device, for example, comprises Si, Ti, and Cr on the cathode electrode 15 is formed. And DLC film 16 is formed using an ECR plasma CVD system. At this time, it exhausted to the degree of vacuum below 1.0×10^{-4}Pa, and as process gas, ethylene was introduced so that it might be set to 0.66 Pa. Microwave power was set to 200W and coil current was set to 16.5A. And DLC film 16 of 500 A of thickness was formed, increasing continuously the RF power impressed to a glass substrate in 0W-120W.

[0029]The organic EL device 10 produced as mentioned above was neglected in the incubator of 85% of humidity at the temperature of 85 ** for 24 hours, and the state of the element was observed with the optical microscope. Neither a crack nor exfoliation of a film was observed in this organic EL device 10, but the result was able to check having gas barrier property and adhesion force with expensive DLC film 16.

[0030]

[Effect of the Invention]According to the manufacturing method of the electroluminescent element concerning this invention, the internal stress distribution of a DLC film can be changed continuously, and the stress concentration to specific lamination interfaces can be prevented. For this reason, while being able to raise the adhesion force of a DLC film and being able to prevent exfoliation of a film, it becomes possible to secure the high gas barrier characteristic. Therefore, invasion of oxygen from the external world or a steam can be prevented certainly, and reinforcement of an element can be attained. The DLC film contains the hydrogen atom.

Since it has spatial room in atomic arrangement, elastic modification is possible.

When this uses the substrate formed with the resin material, while being able to make bending free, the damage to a crack, exfoliation, etc. does not arise in the DLC film as a protective film with the stress added at the time of bending.

[Translation done.]

CLAIMS

[Claim(s)]

[Claim 1]It faces forming a DLC (Diamond Like Carbon) film as a protective film with plasma CVD method on an electroluminescent element by which thin film forming was carried out on a substrate, A manufacturing method of an electroluminescent element controlling to become large as RF power impressed to the above-mentioned substrate is increased continuously and internal stress distribution of the DLC film concerned goes in a membrane surface from an inside of a film during membrane formation of the above-mentioned DLC film.

[Claim 2]A manufacturing method of the electroluminescent element according to claim 1 carrying out variation width of RF power impressed to the above-mentioned substrate within the limits of conditions from which the above-mentioned DLC film serves as a sp3 joint subject with a maximum RF power value.

[Claim 3]A manufacturing method of the electroluminescent element according to claim 1 setting to 120W the maximum of RF power impressed to the above